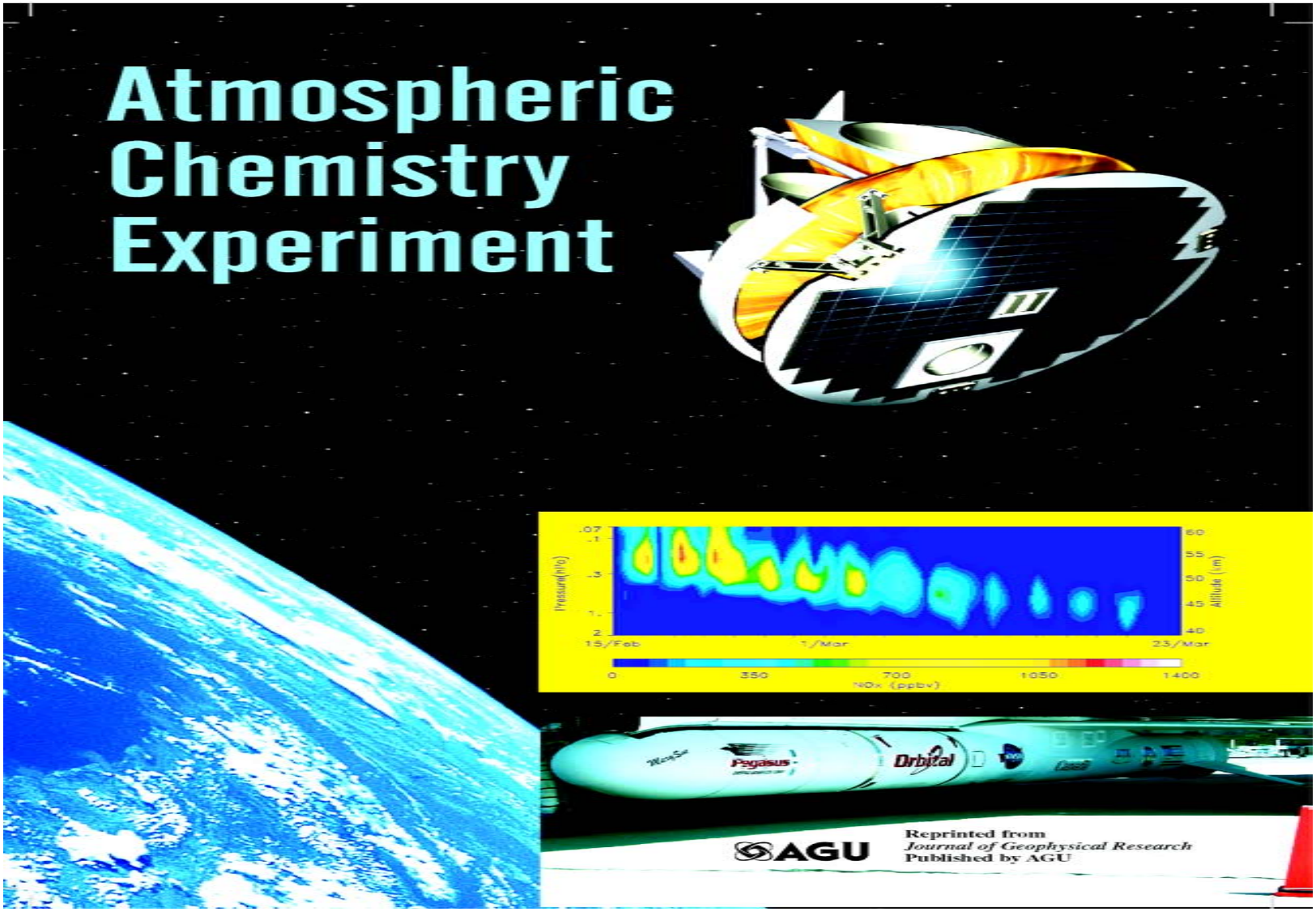
Upper Tropospheric Measurements of Biomass Burning Emissions with the Atmospheric Chemistry Experiment (ACE) Fourier Transform Spectrometer



ACE Mission and Status

- ACE is a Canadian-lead mission successfully launched into a 74º inclined orbit by a U.S.-supplied Pegasus launch vehicle on August 12, 2003
- The primary instrument is a Fourier transform spectrometer (FTS) operating in solar occultation mode
- Additional measurements are obtained in the visible with arrays and a UV-Visible spectrometer
- FTS spectral coverage of 750-4500 cm^{-1} often with measurements of multiple bands for a molecule providing consistency tests of spectroscopic parameters
- Spectra are recorded with a maximum optical depth of 25 cm and a scan time of 2 s and provide 3-4 km vertical resolution
- Instrument is continuing to operate well and has provided an order of magnitude more measurements than ATMOS during its four shuttle flights

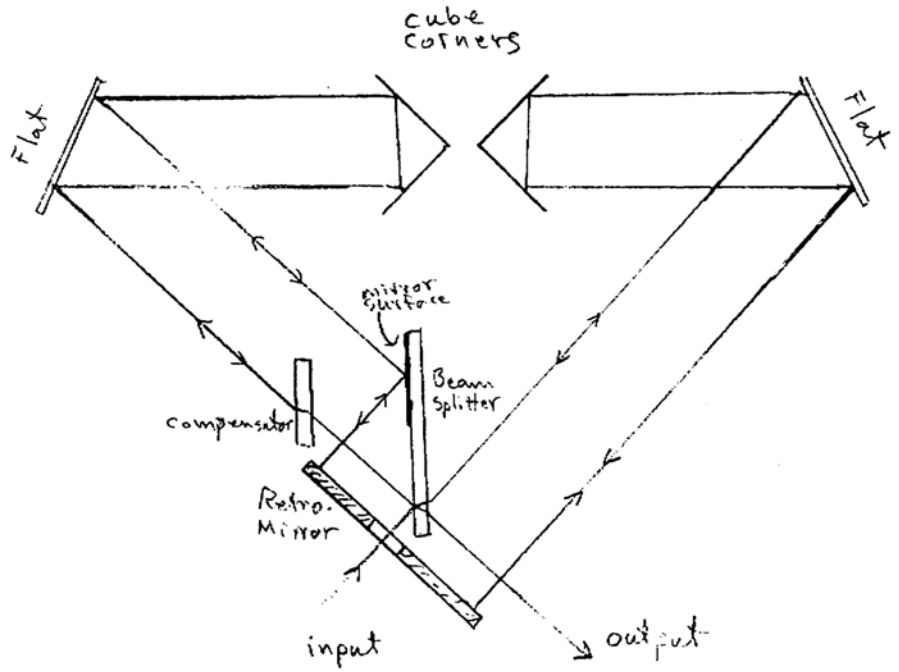
Instruments

- Infrared Fourier Transform Spectrometer operating between 2 and 13 microns with a resolution of 0.02 cm^{-1}
- 2-channel visible/near infrared Imagers, operating at 0.525 and 1.02 microns (cf., SAGE II)
- Suntracker keeps the instruments pointed at the sun's radiometric center.
- UV / Visible spectrometer (MAESTRO) 0.285 to 1.03 microns, resolution ~1-2 nm
- Startracker

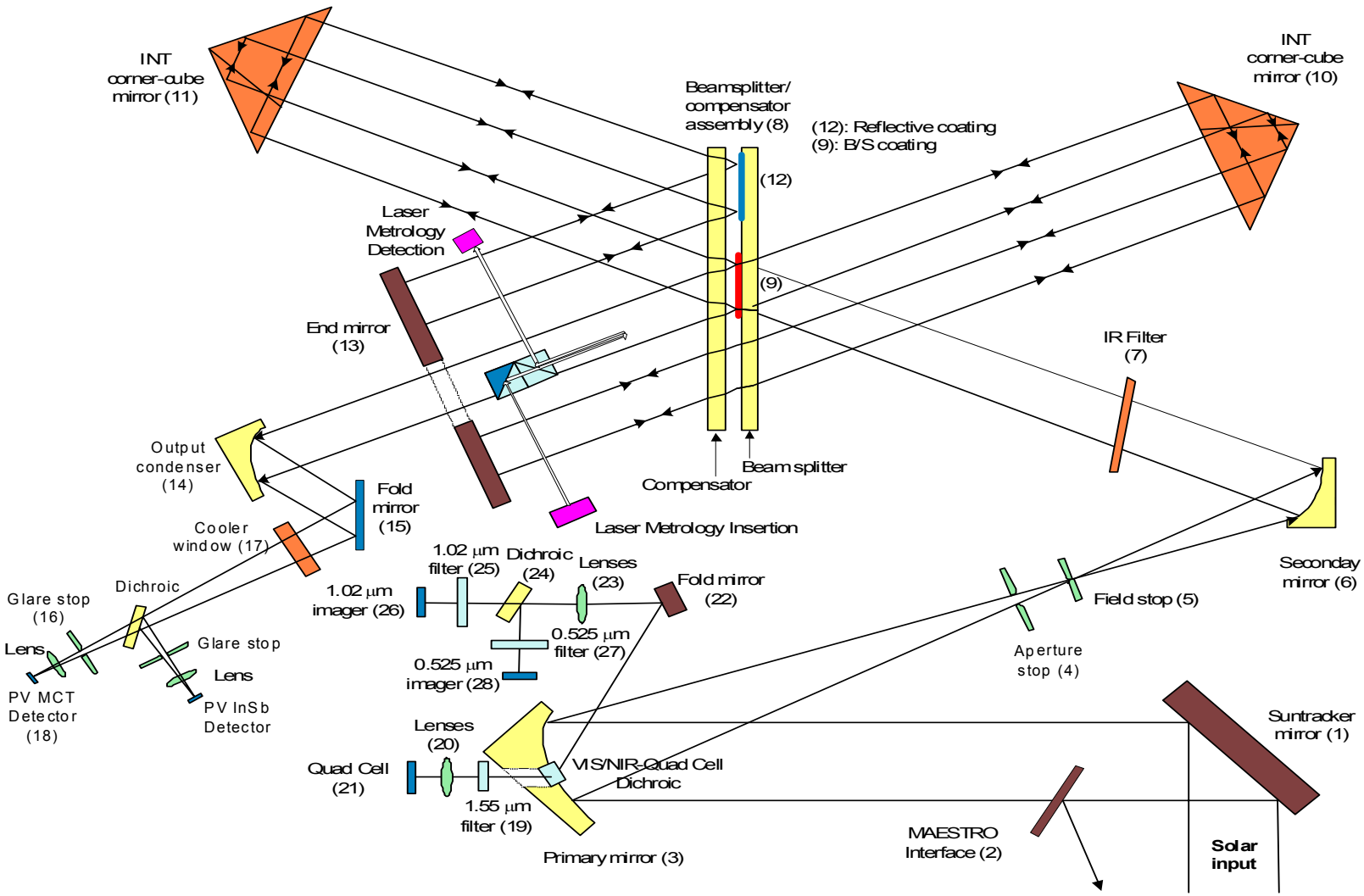
The Concept

Jennings
7 Jan 98

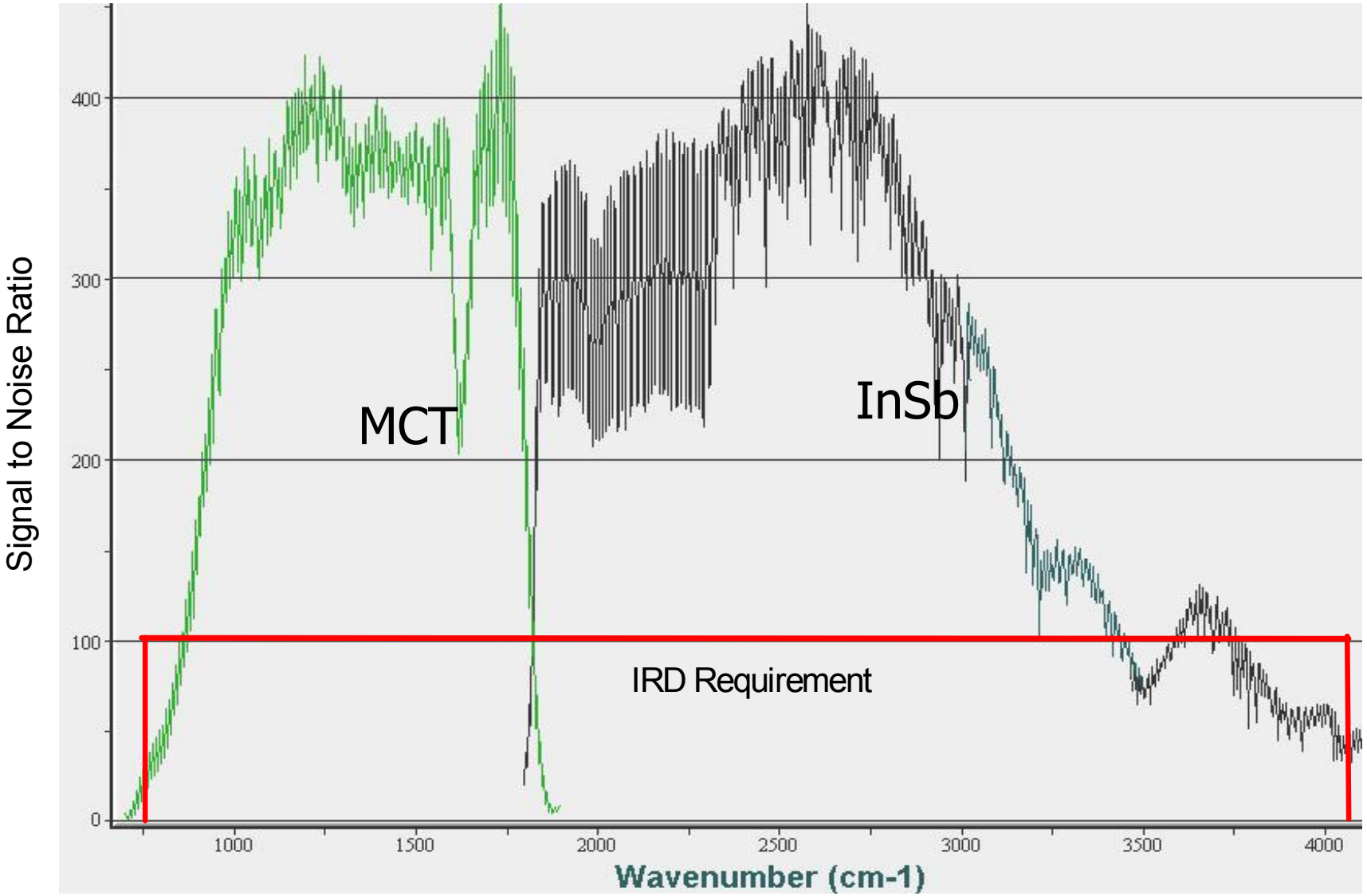
Web Concept



Optical Layout (ABB-Bomem)



SNR Performance

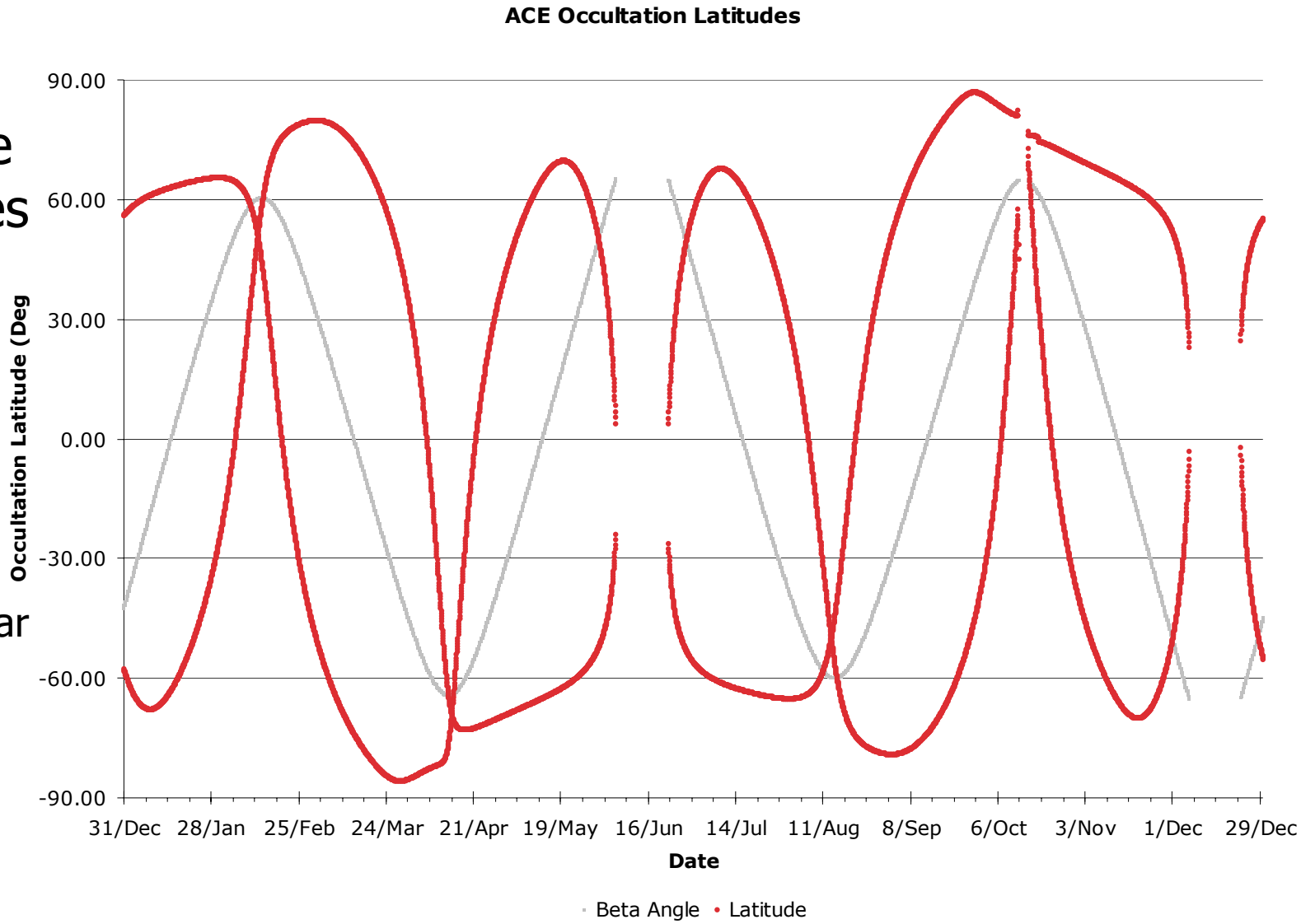


Successful Pegasus XL Launch
Aug. 12, 2003-Vandenberg AFB

Global Coverage

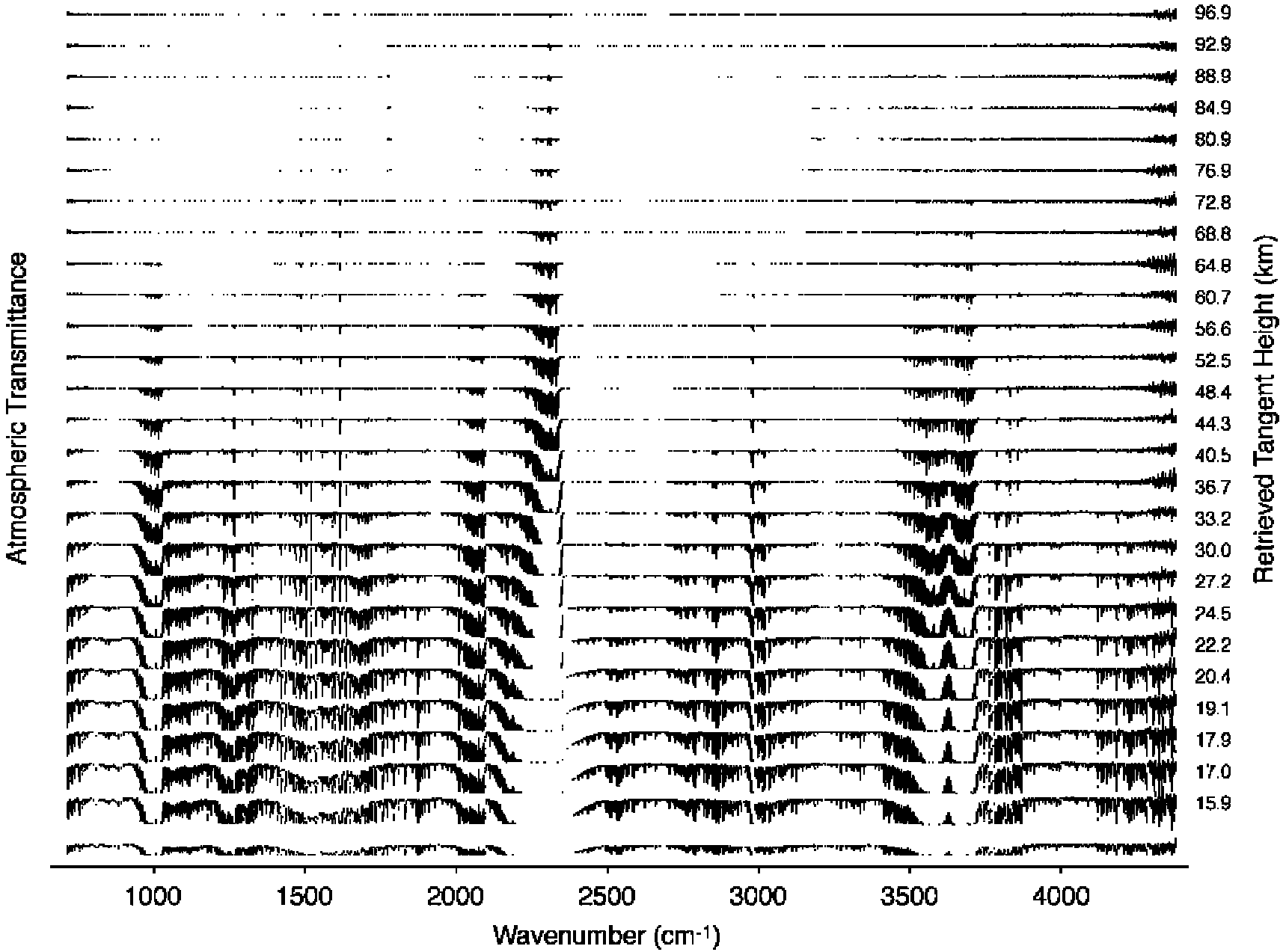
Latitude /degrees

650 km, 74° inclined circular orbit, RAAN 55.7°

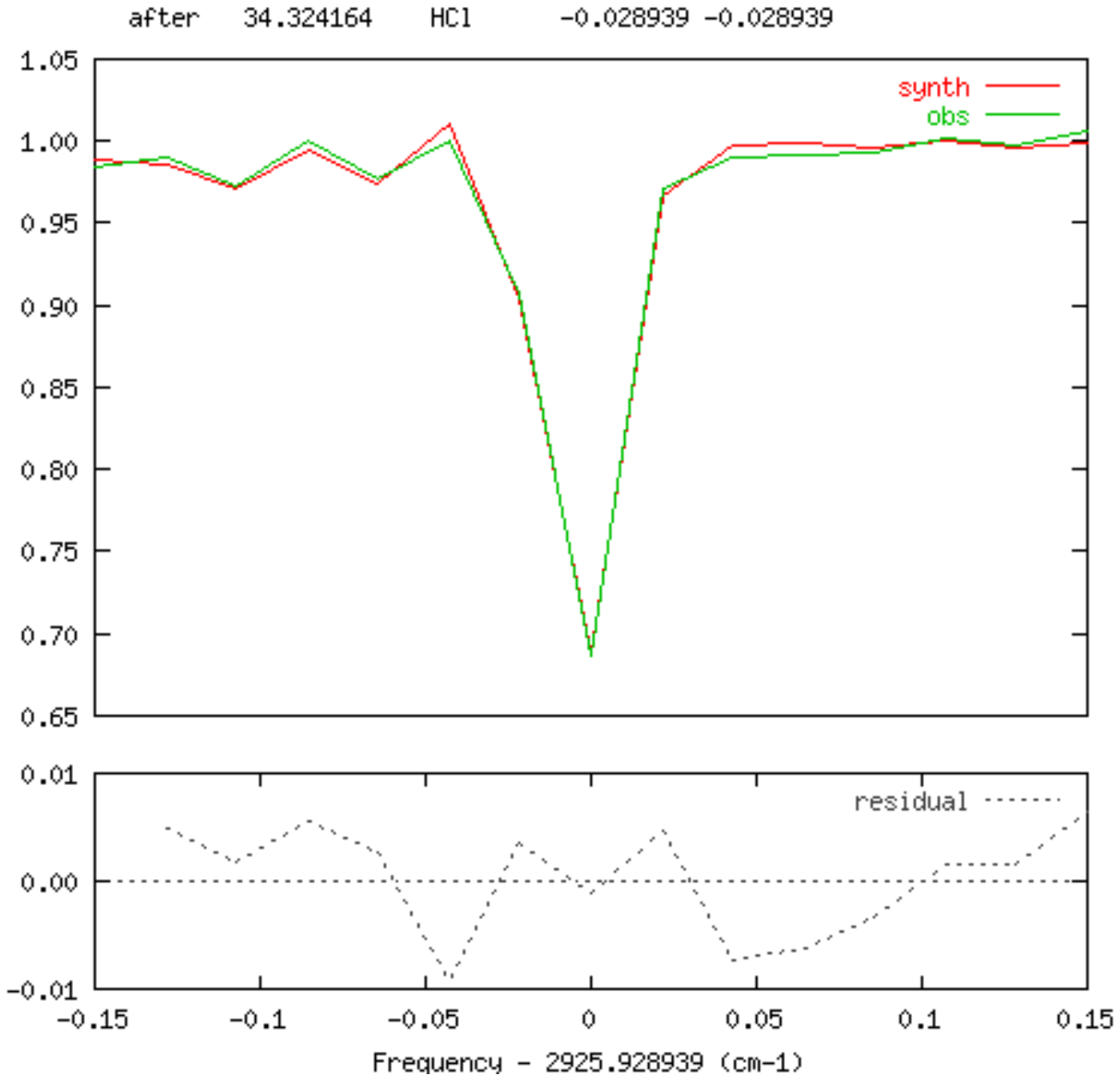


Jan. 1, 2004 to Dec. 29, 2004

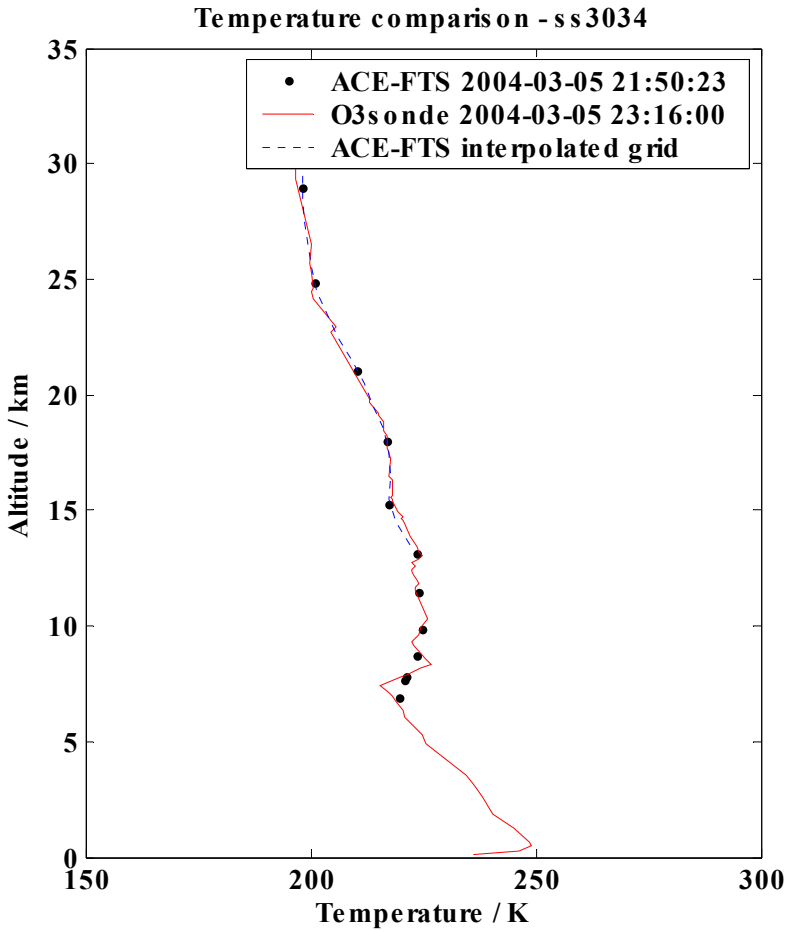
Occultation sequence

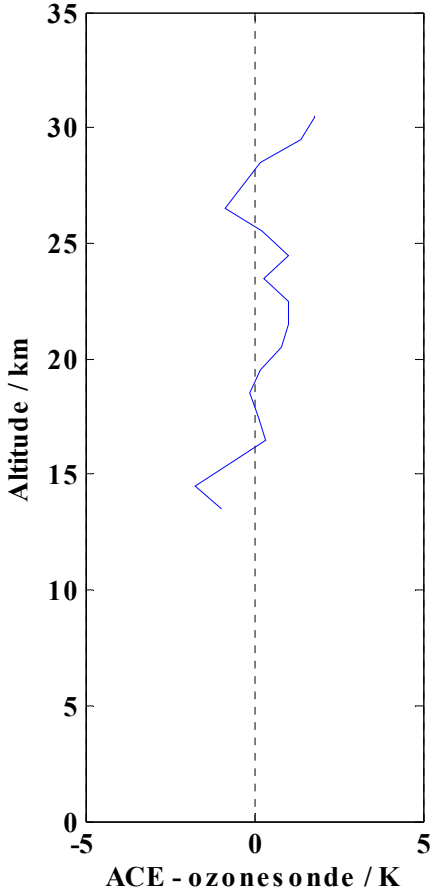


HCl R(0) at 34 km



ACE-Radiosonde Temperature Comparison (Eureka)



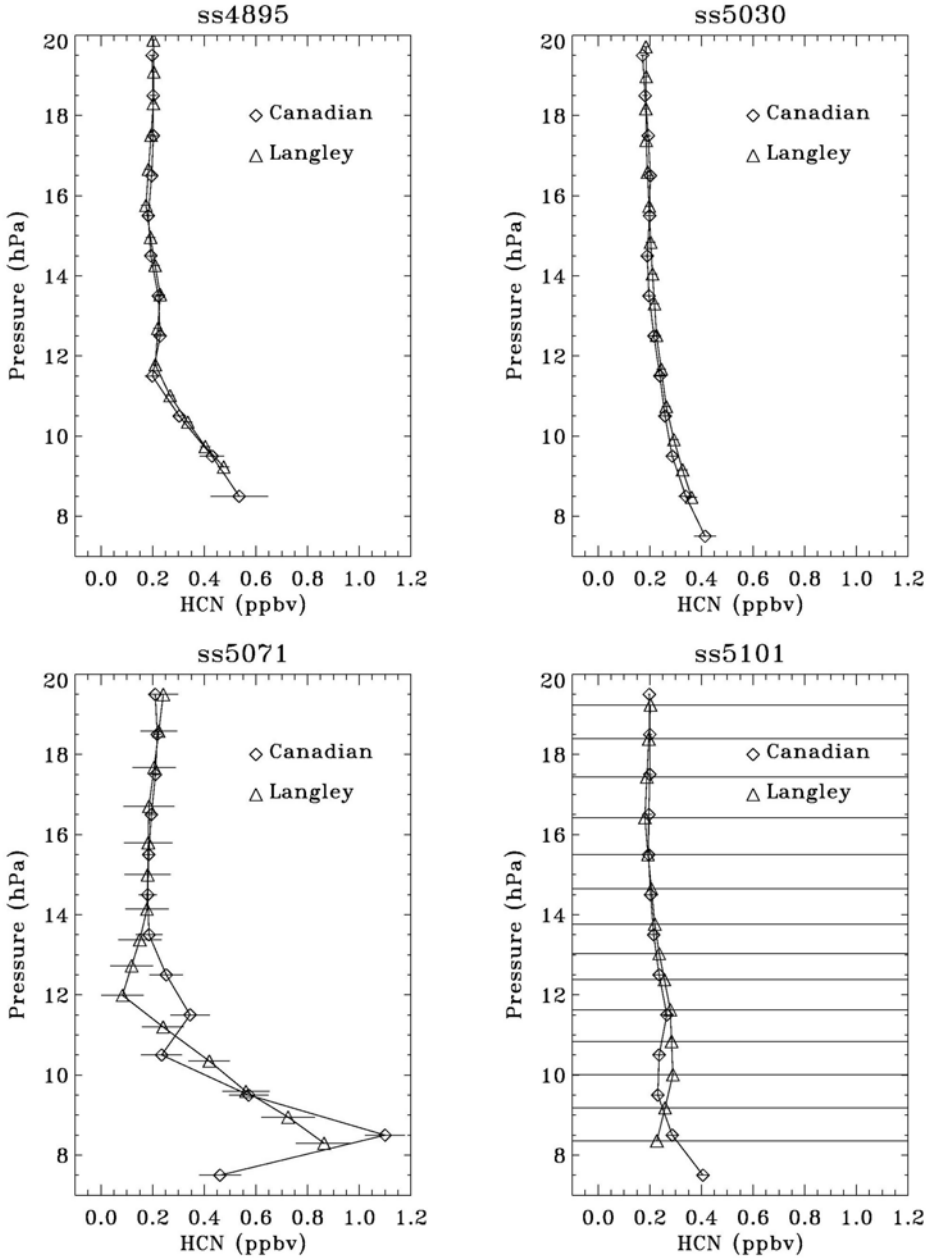


Maximum difference
15-25 km: 1 K
Overall: 1.8 K

For 8 profiles – within 200 km of Eureka
Max. mean diff. = 1.7 K

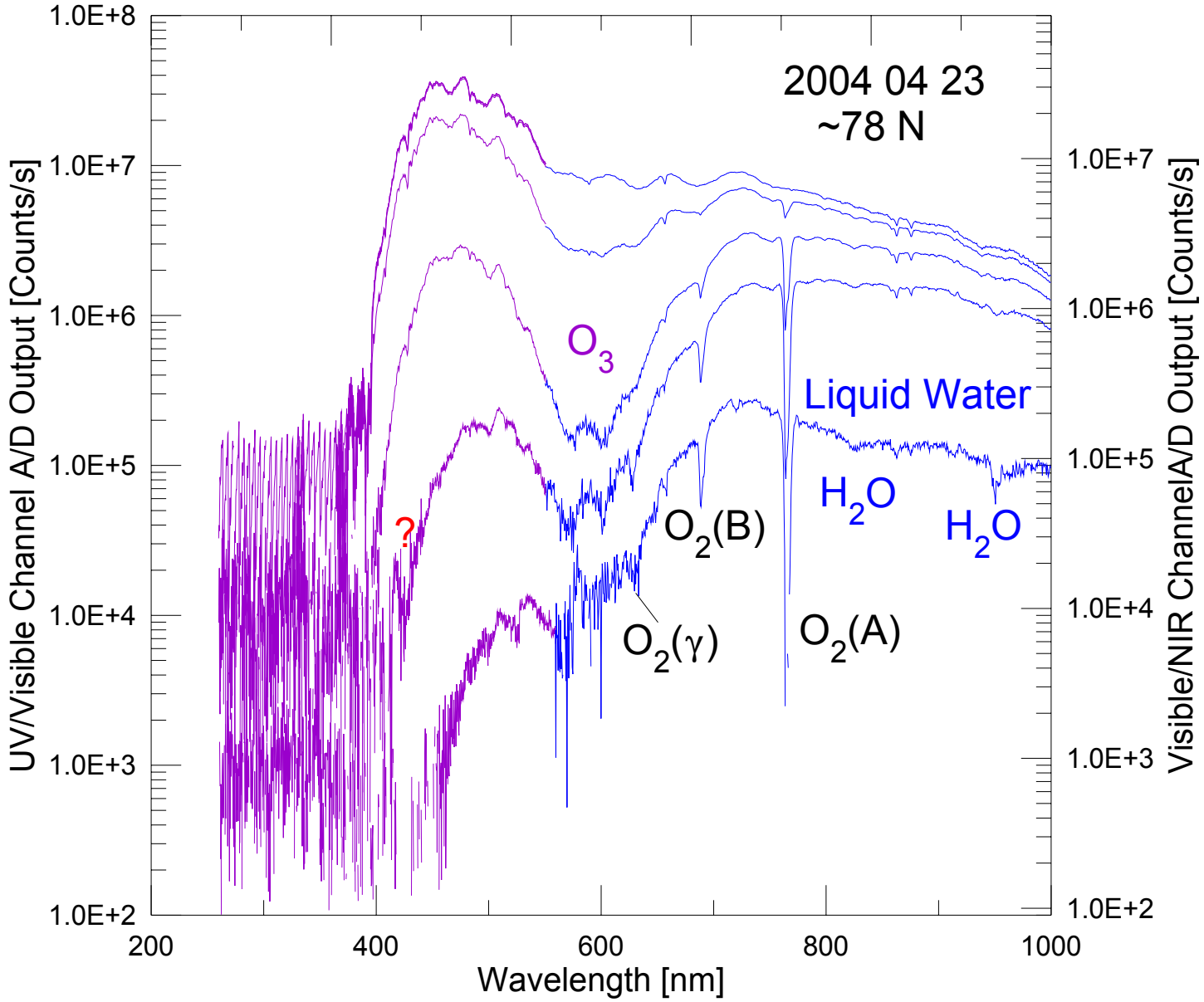
Will be making comparisons with CHAMP, SABER and lidar measurements

HCN-Langley and Waterloo



Rinsland et al.

MAESTRO Spectra



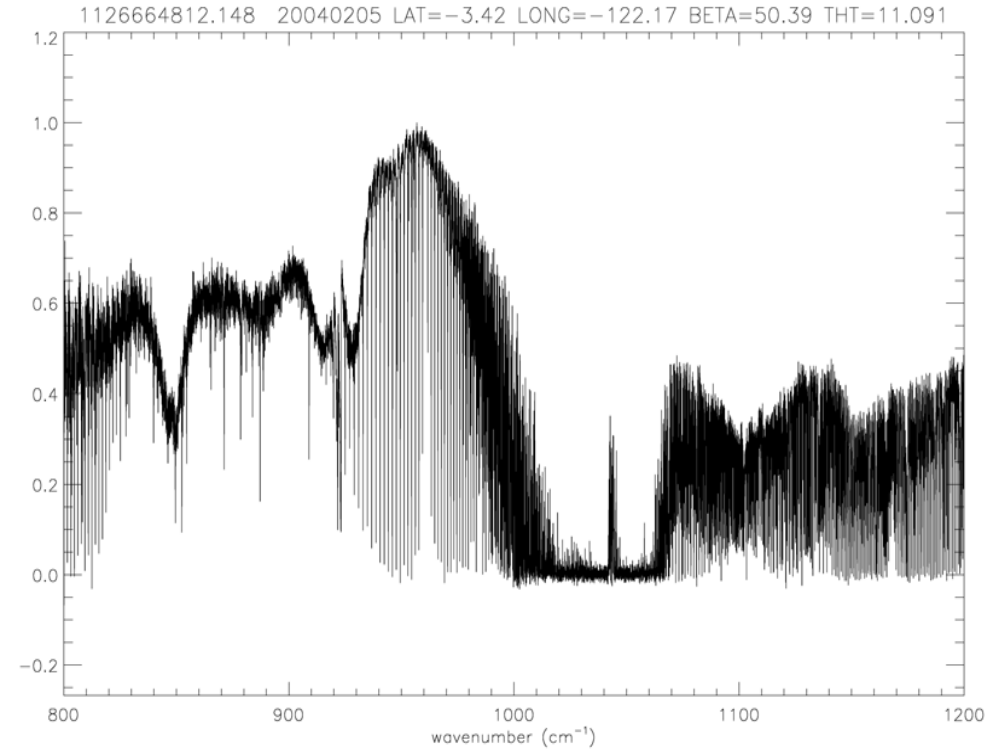
Standard Products

- H2O
- O3
- N2O
- CO
- CH4
- NO
- NO2
- HNO3
- HF
- HCL
- CLO
- OCS
- HOCL
- H2O2
- HO2NO2
- N2O5
- CLONO2
- HCN
- CH3CL
- CF4
- CCL2F2
- CCL3F
- CCL4
- COF2
- C2H6
- C2H2
- N2
- CHF2CL
- HCOOH
- SF6
- H2O(181)
- H2O(171)
- H2O(162)
- Temperature

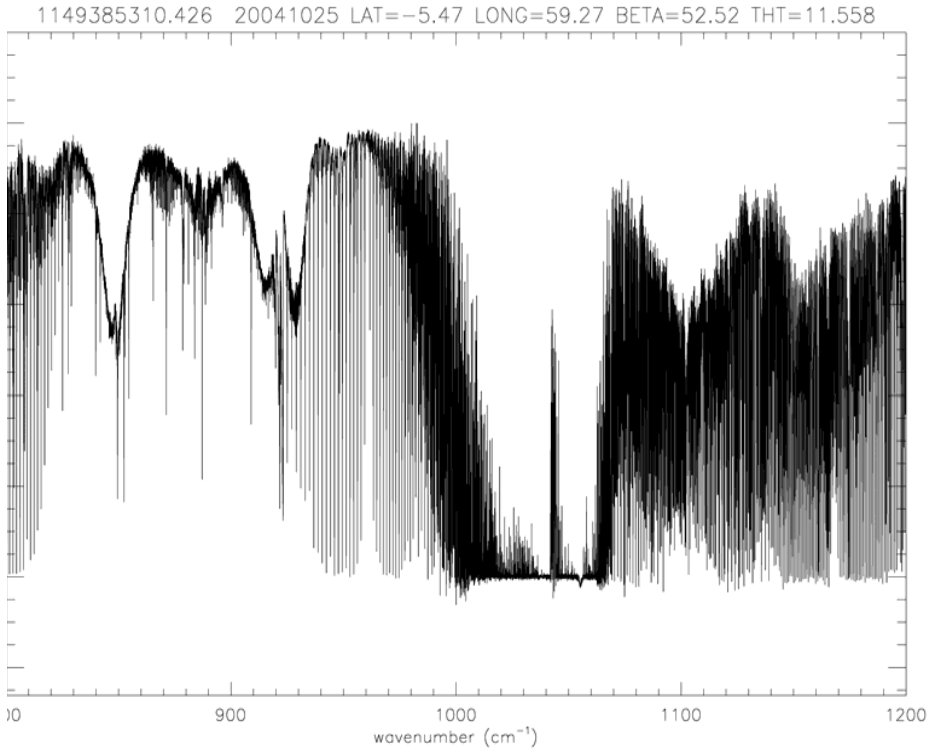
ACE Molecular Detections

- CH_3OH (Methanol)
  - Dufour et al. ACP, 6, 3463-3470, 2006
  - Most abundant organic molecule after CH_4 in the troposphere
- HFC134a
- HCOOH
- CFC-113
- COCIF

Tropical Occultations near 11 km with and without Clouds

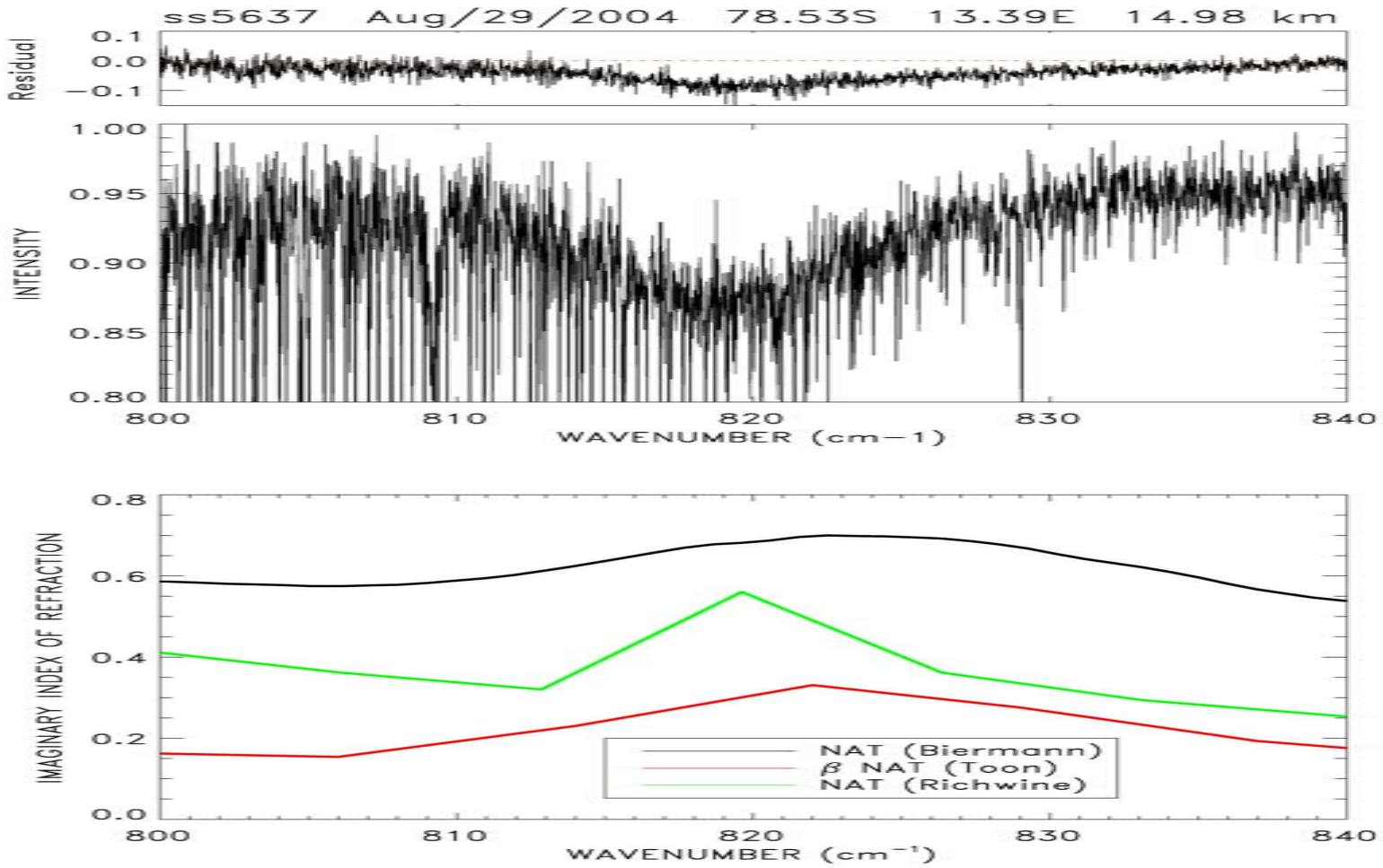


ss2549

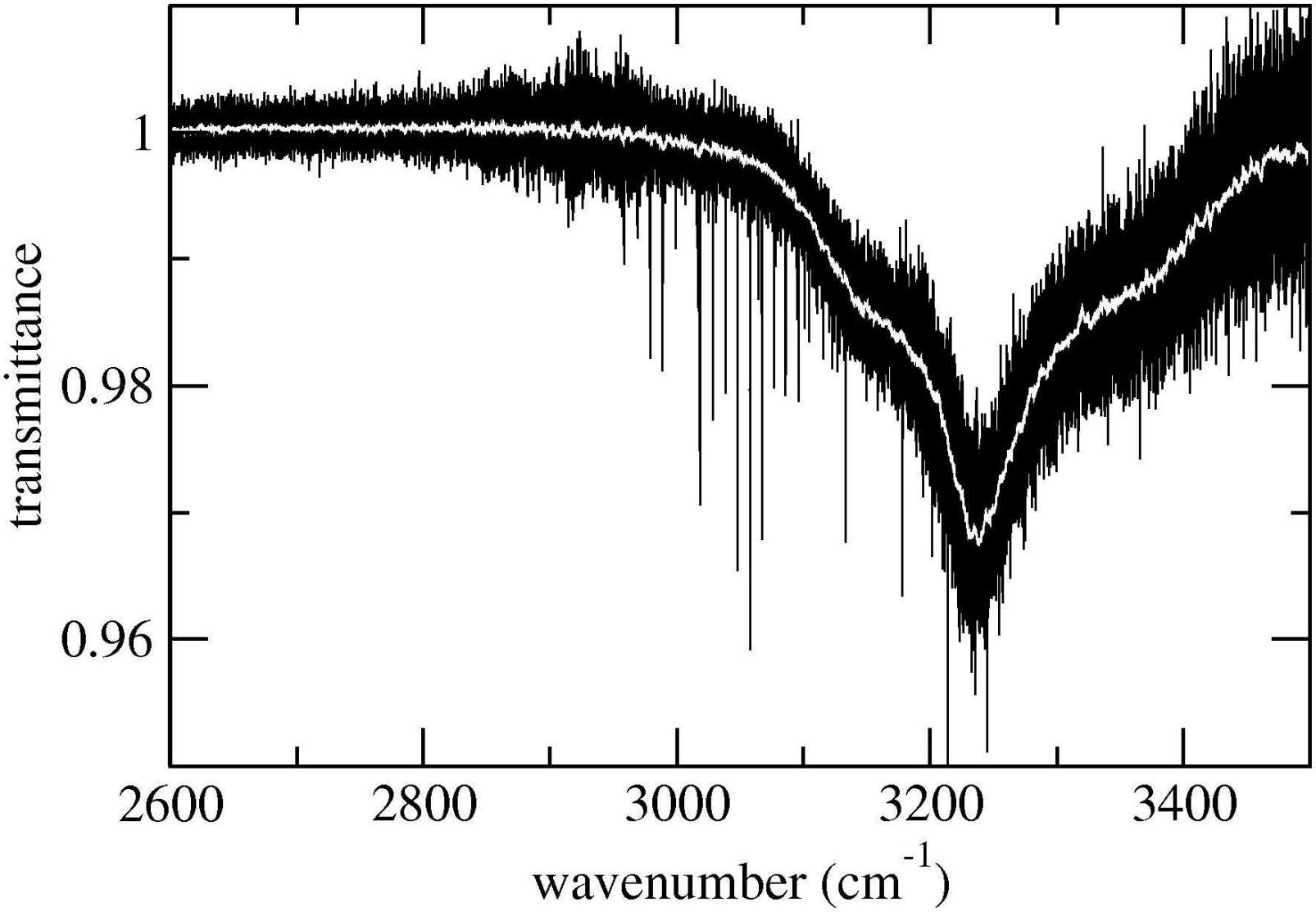


sr6470

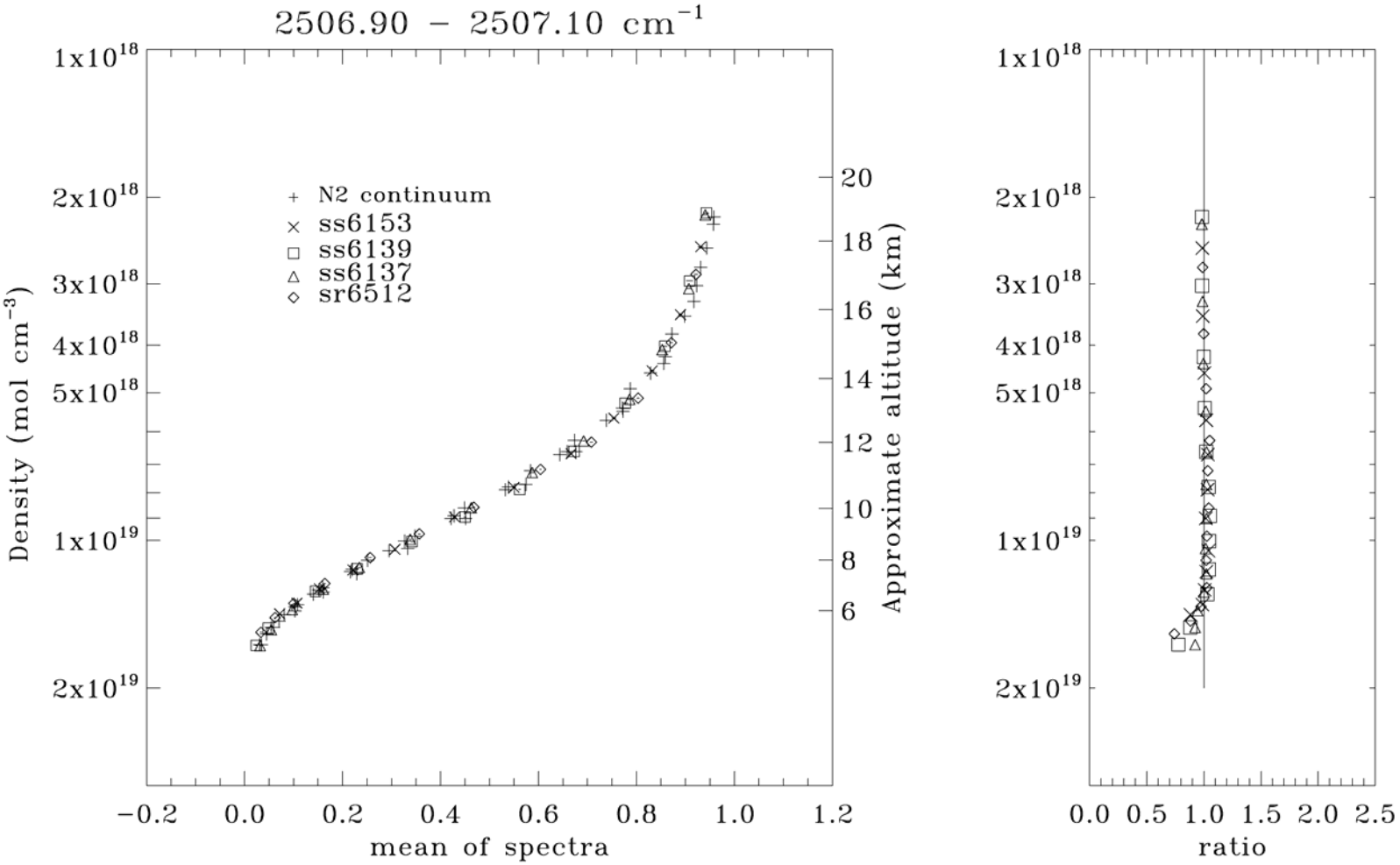
ACE ss5637


ss5637 Aug/29/2004 78.53S 13.39E 14.98 km
Residual
0.1
0.0
−0.1
INTENSITY
1.00
0.95
0.90
0.85
0.80
800
810
820
830
840
WAVENUMBER (cm−1)
IMAGINARY INDEX OF REFRACTION
0.8
0.6
0.4
0.2
0.0
NAT (Biermann)
β NAT (Toon)
NAT (Richwine)
800
810
820
830
840
WAVENUMBER (cm⁻¹)

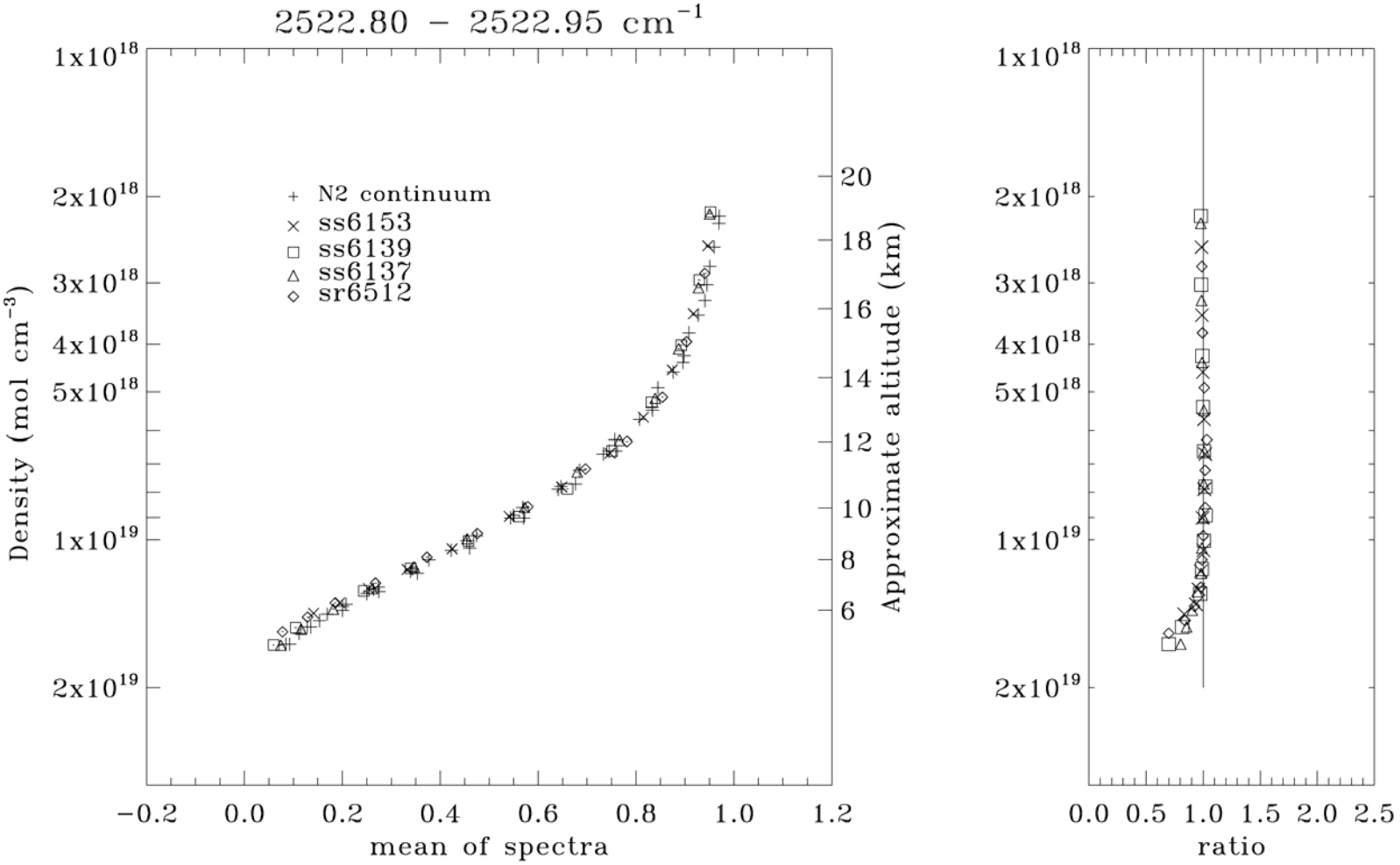
ACE Spectra of Polar Mesospheric Clouds (PMCs)



Precise Determination of Density from N_2 Continuum Extinction



Precise Determination of Density from N_2 Continuum Extinction



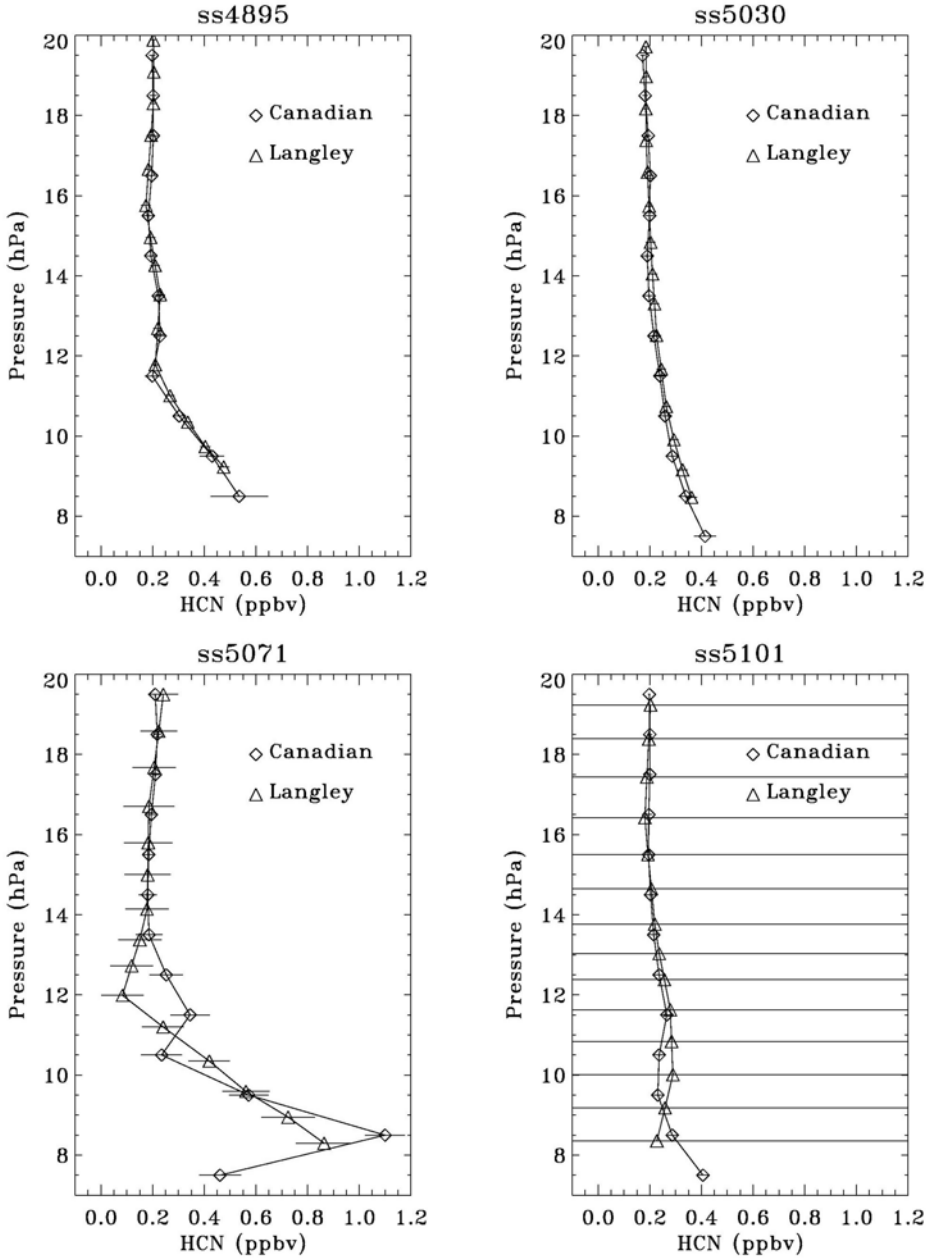
ACE Tropical and Southern Mid-Latitude Measurements

- The ACE orbit samples the tropics and mid-latitudes of the southern hemisphere only for a brief period each year
- The measurements coincide with the dry season of peak biomass burning in the tropics
- Previous NDSC studies have shown the tropical emissions produce elevated biomass burning products (CO, C_2H_6, HCN, C_2H_2) with lifetimes long enough to be readily measured with high resolution infrared instruments
- MOPITT measurements also show impact from those fires on CO and aerosols [Edwards et al. 2006]

Tropospheric Microwindows for CO

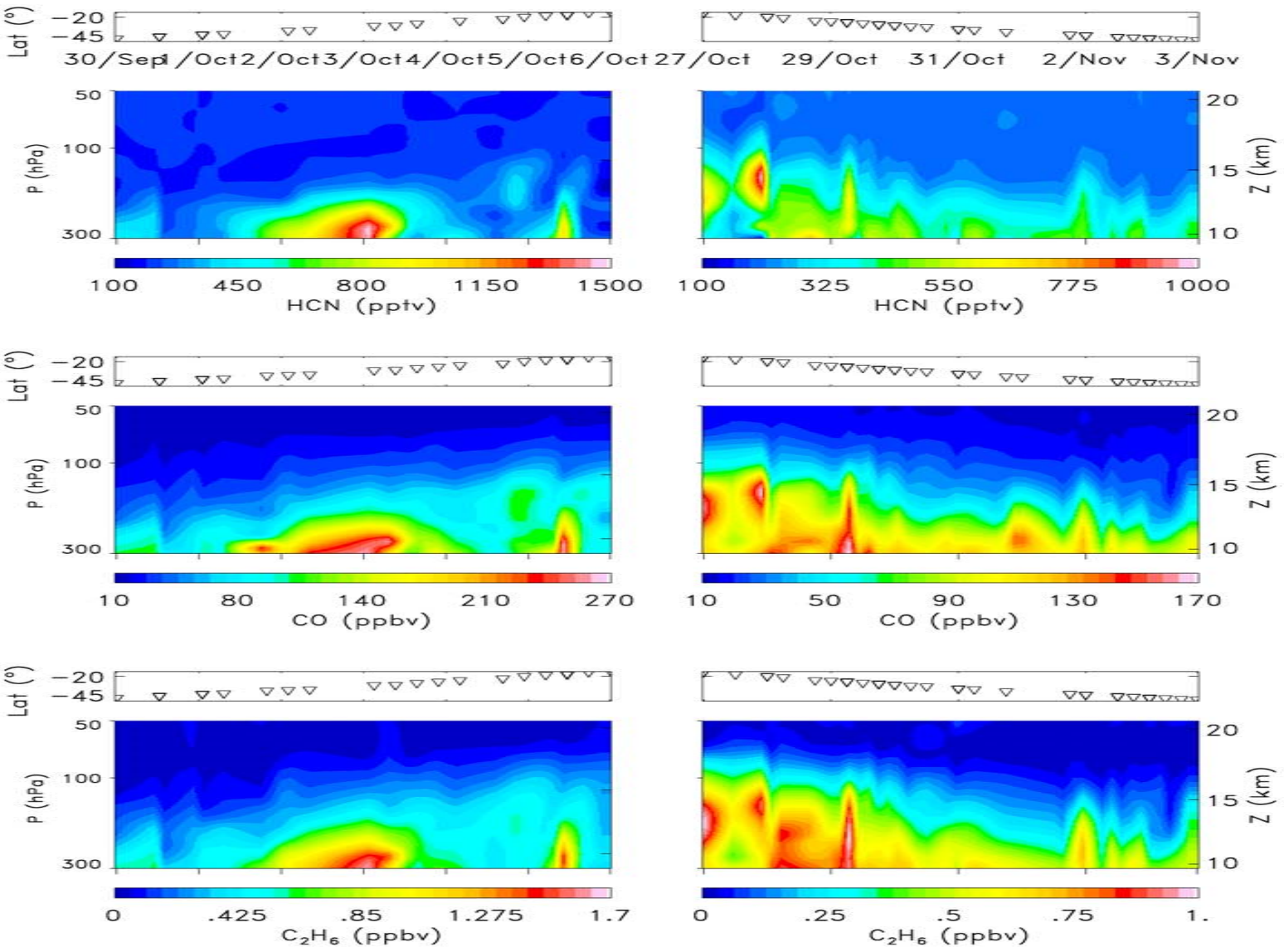
Microwindow	Gas	Altitude Range	
		Low	High
4209.20 - 4209.55	CO	4.0	15.0
	CH4	4.0	40.0
4222.48 - 4223.28	CO	4.0	15.0
	CH4	20.0	40.0
4226.65 - 4227.70	CO	4.0	15.0
	CH4	4.0	40.0
4235.75 - 4236.30	CO	5.0	15.0
	CH4	5.0	40.0
4248.10 - 4248.59	CO	4.0	15.0
	CH4	4.0	40.0
4274.55 - 4274.90	CO	4.0	15.0
	CH4	20.0	45.0
4277.50 - 4279.00	CO	4.0	25.0
	CH4	4.0	45.0

HCN-Langley and Waterloo

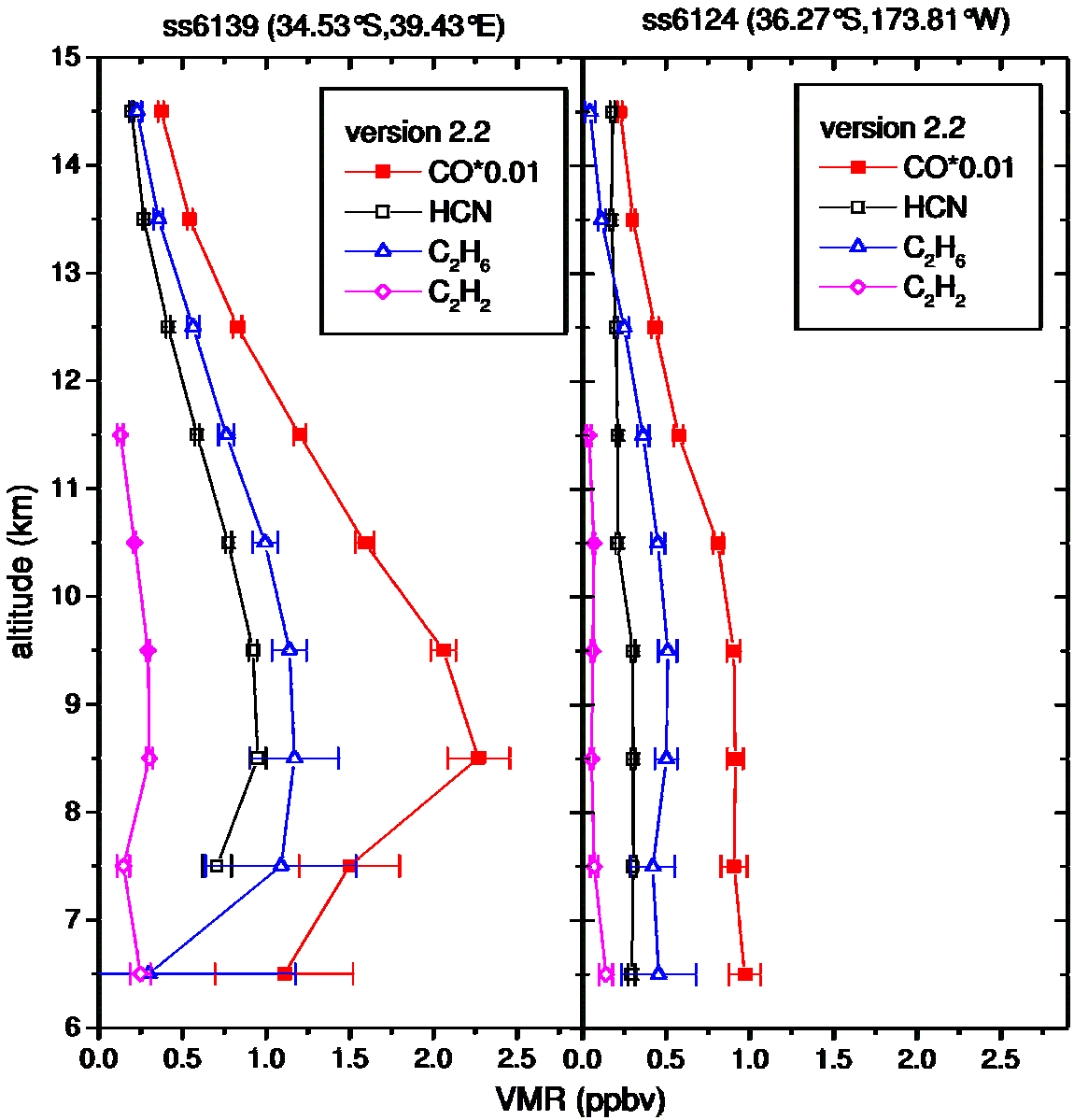


Rinsland et al.

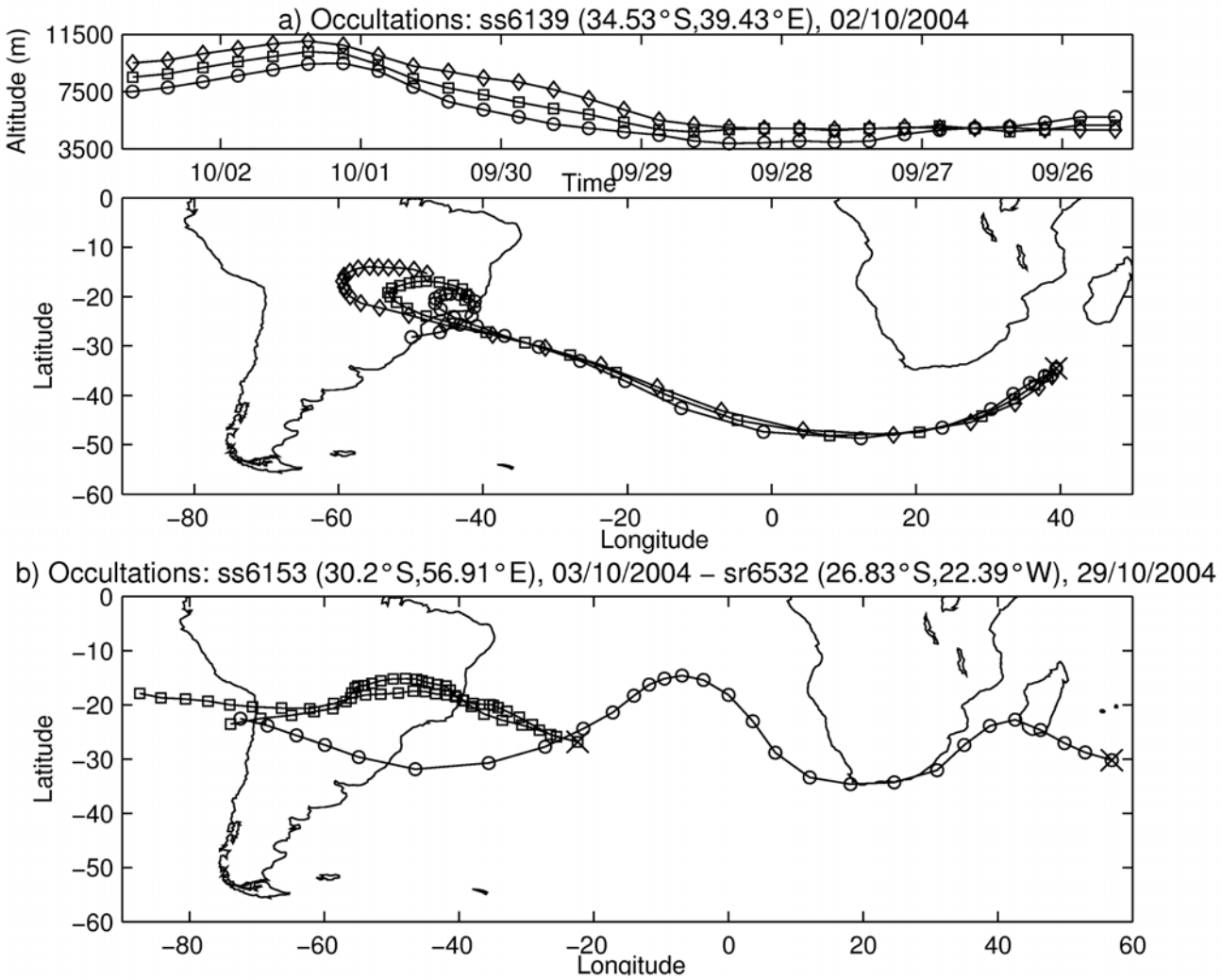
ACE HCN, CO, and C_2H_6 Tropical Biomass Burning Measurements



Sample ACE Enhanced and Background Mixing Ratios from the 2004 Fire Period



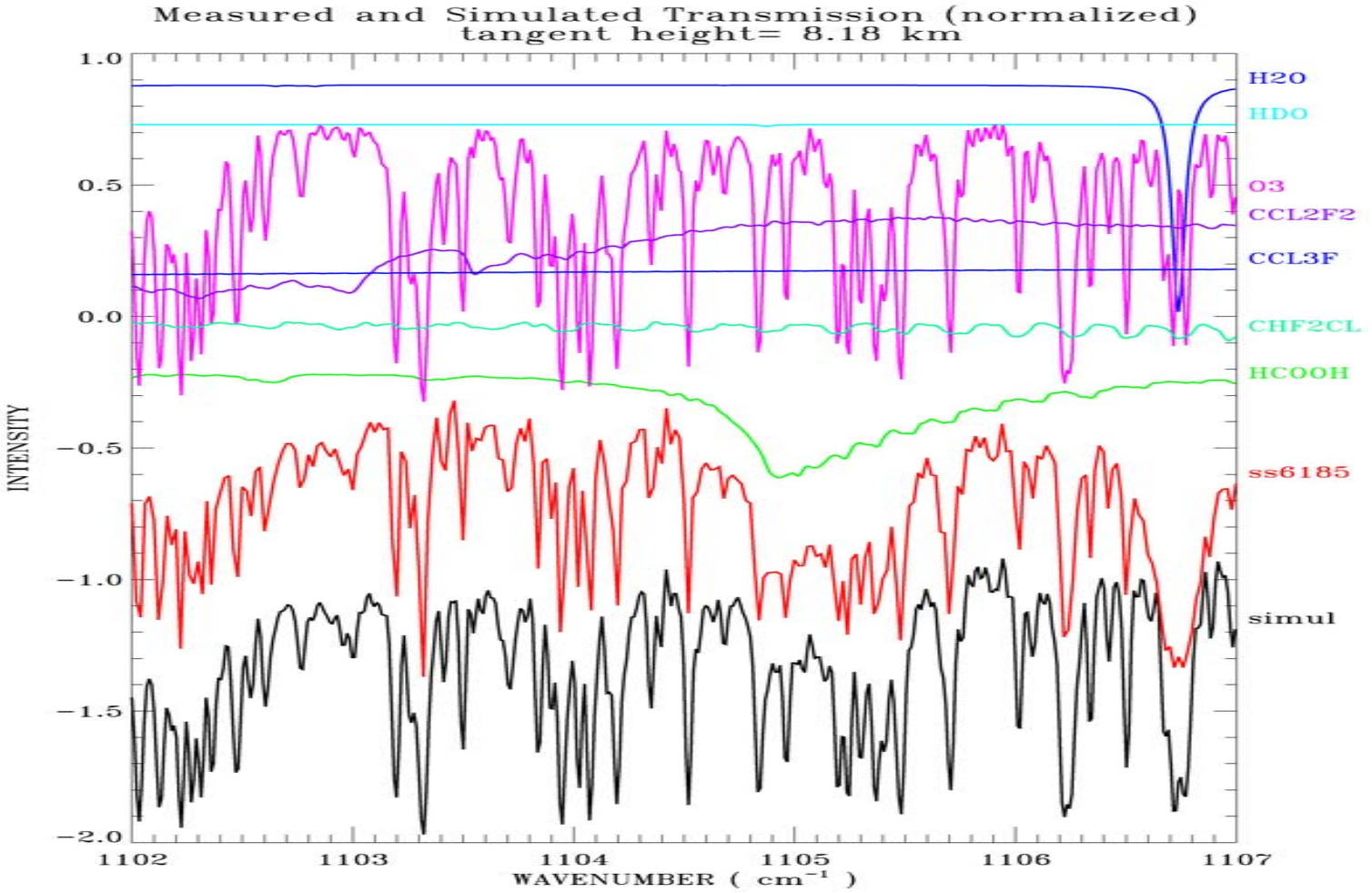
Back Trajectory Calculations



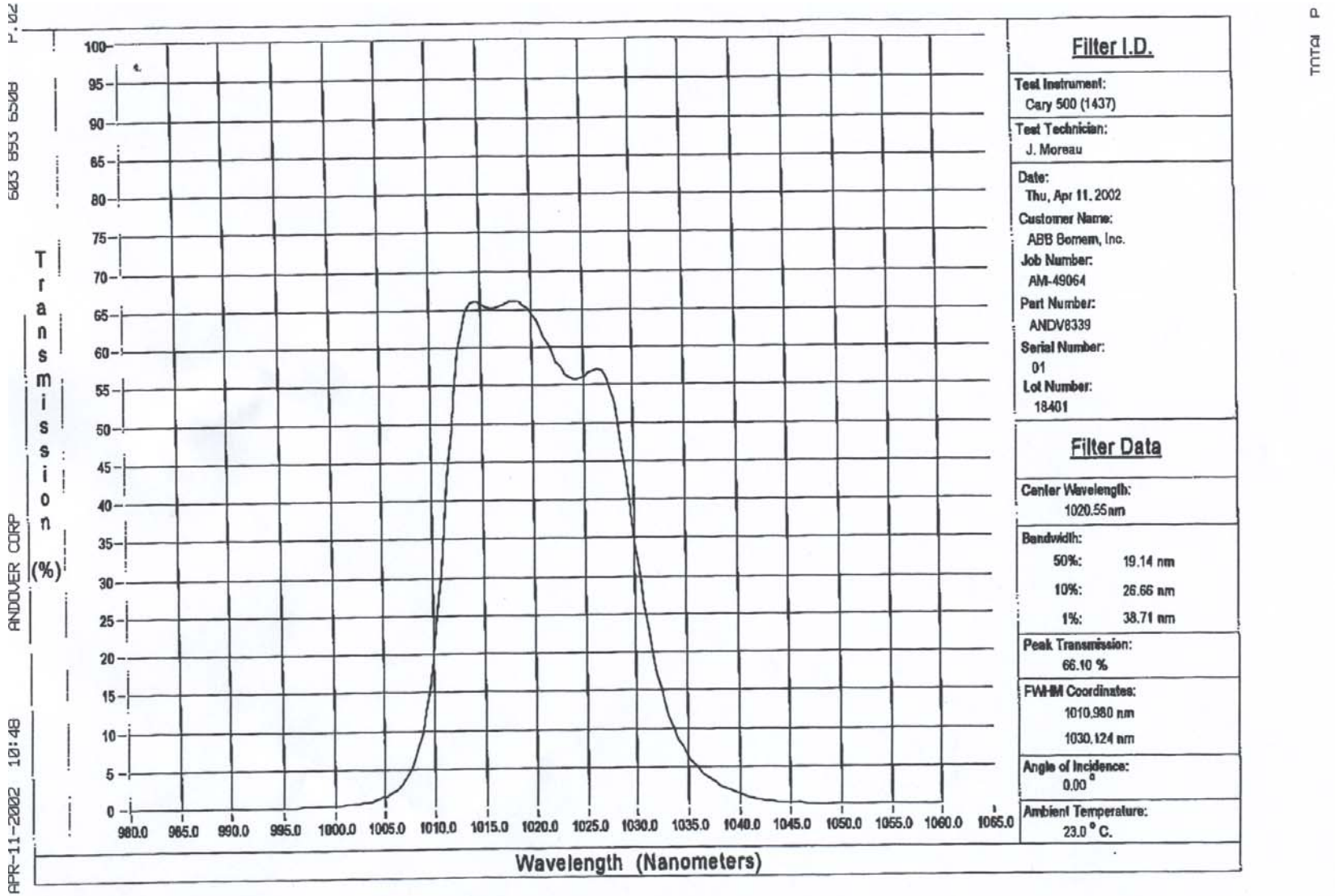
Formic Acid (HCOOH)

- HCOOH has been observed throughout the troposphere
- It is an important oxygenated volatile organic compound (OVOC) with major limitations recognized in the ability of models to reproduce simultaneous measurements of OVOC chemistry, particularly in the dry upper troposphere where OVOCs are a major source of HO_x ($OH+HO_2$) in the background troposphere
  - Sources
    - Biomass burning
    - Biogenic emissions from vegetation
    - Secondary production from organic precursors
    - Motor vehicle emissions
  - Sinks
    - OH reactions in cloud

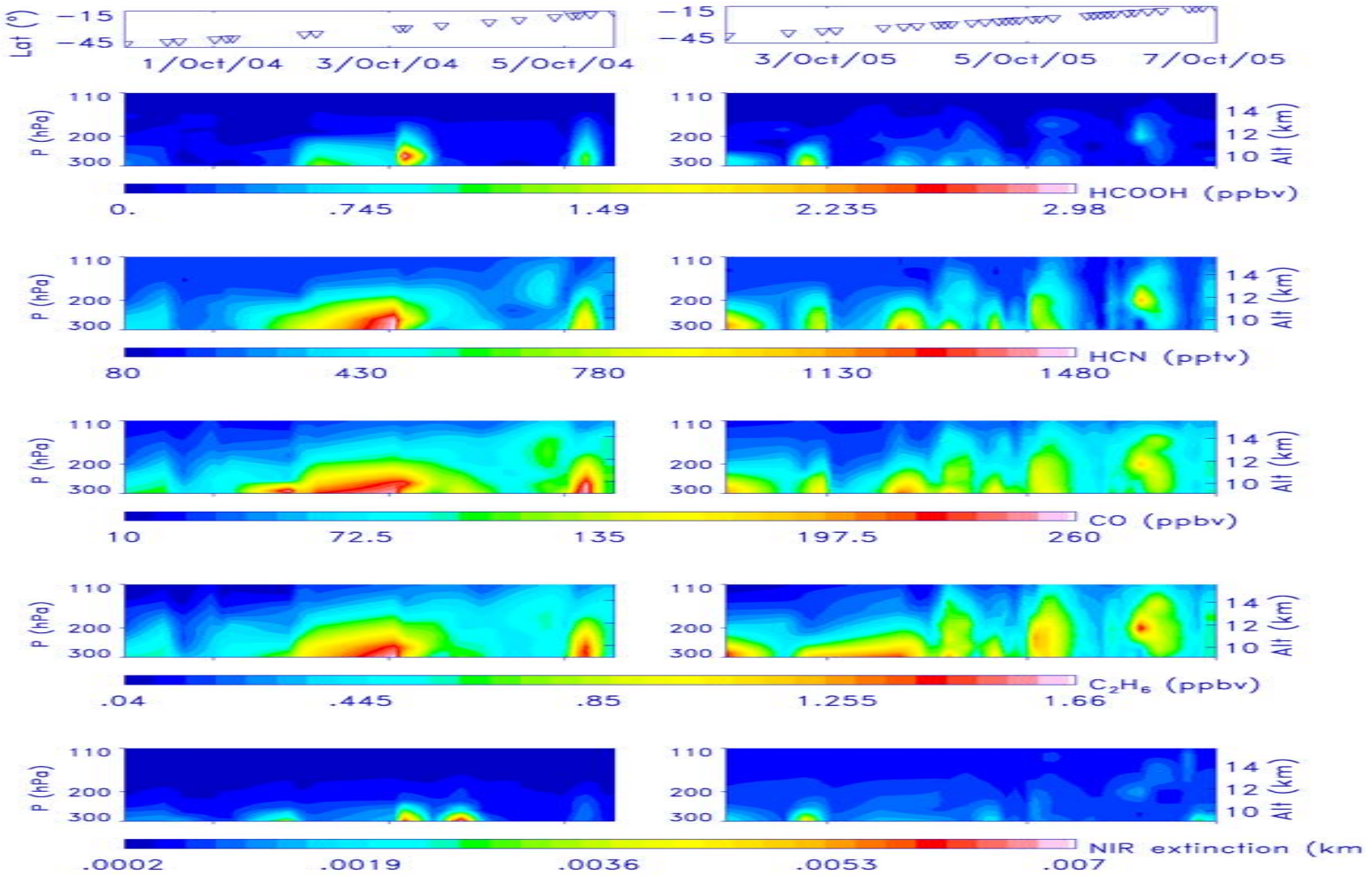
Simulated and ACE Upper Tropospheric Spectra near HCOOH v_6 Band Q branch



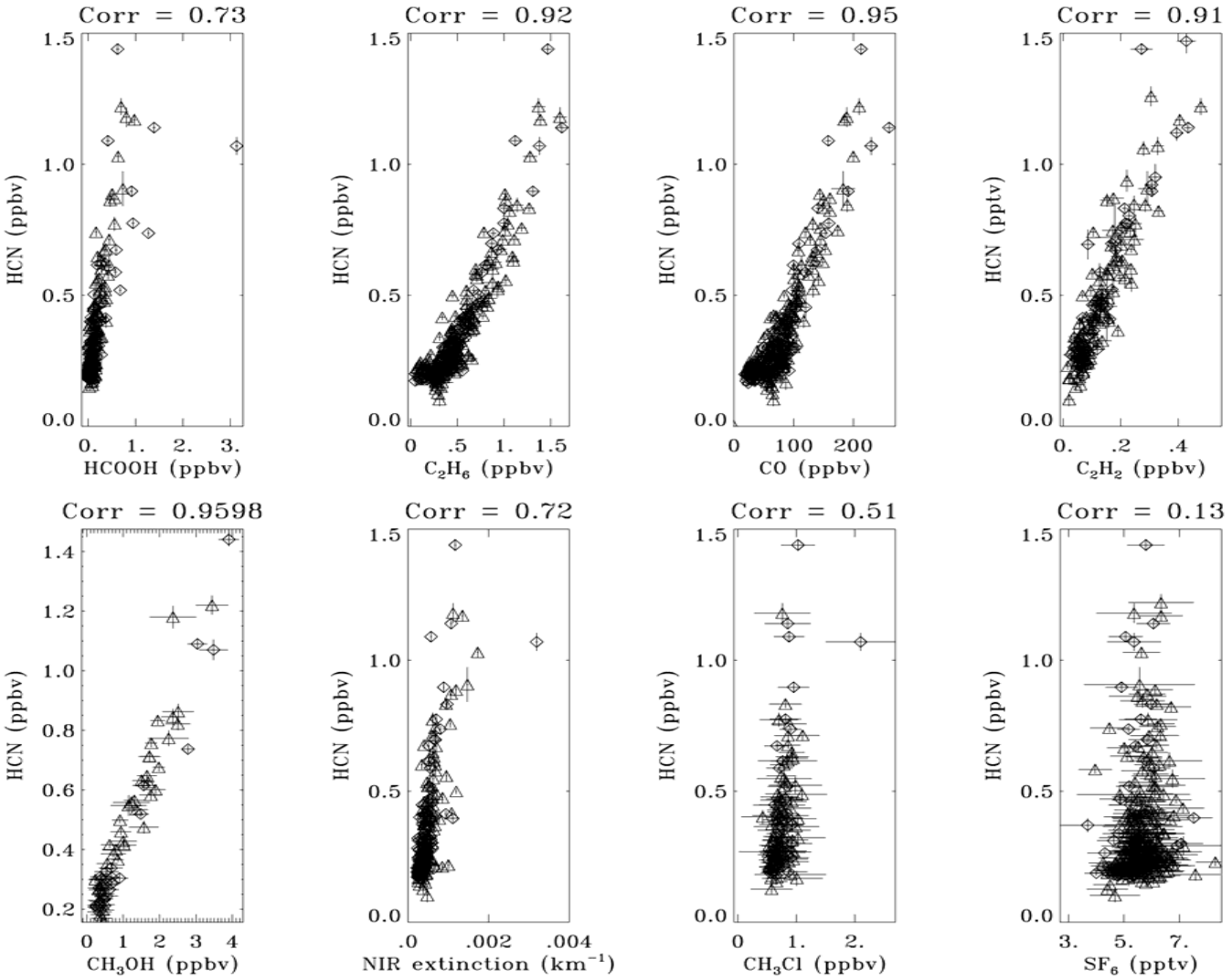
ACE Near-Infrared Imager Filter Curve



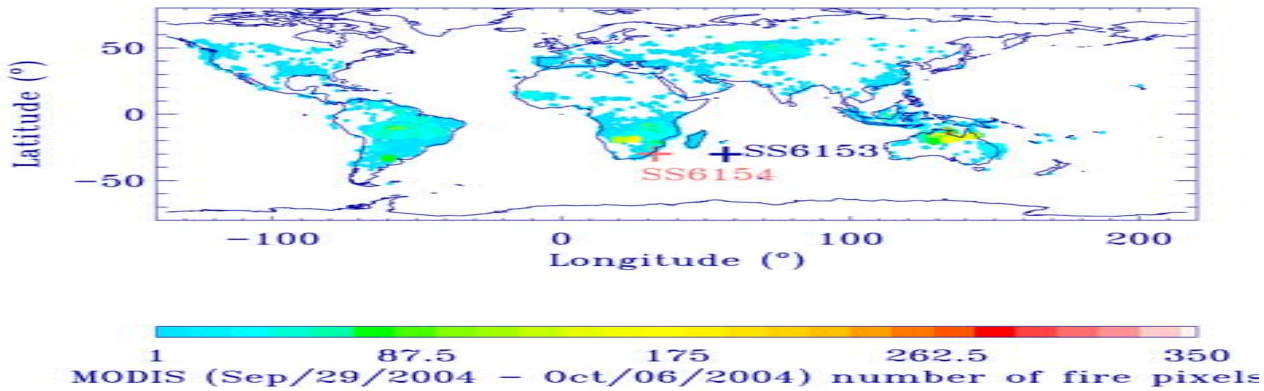
ACE Southern Hemisphere HCOOH Time Series

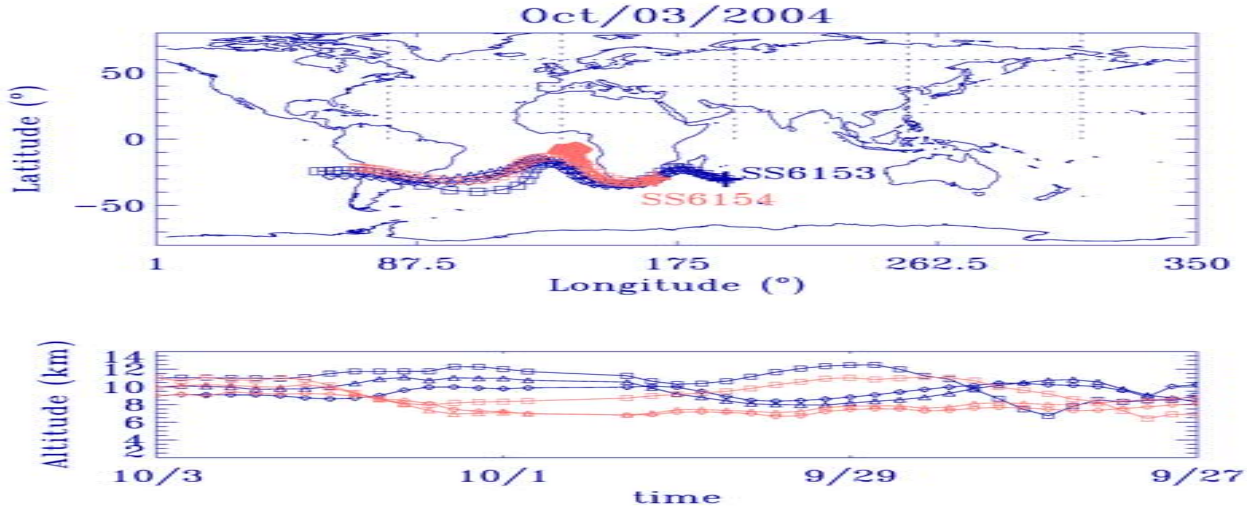


Correlations with HCN



MODIS Fire Counts and Back Trajectories for ss6153 and ss6154





Impact of Boreal Biomass Burning

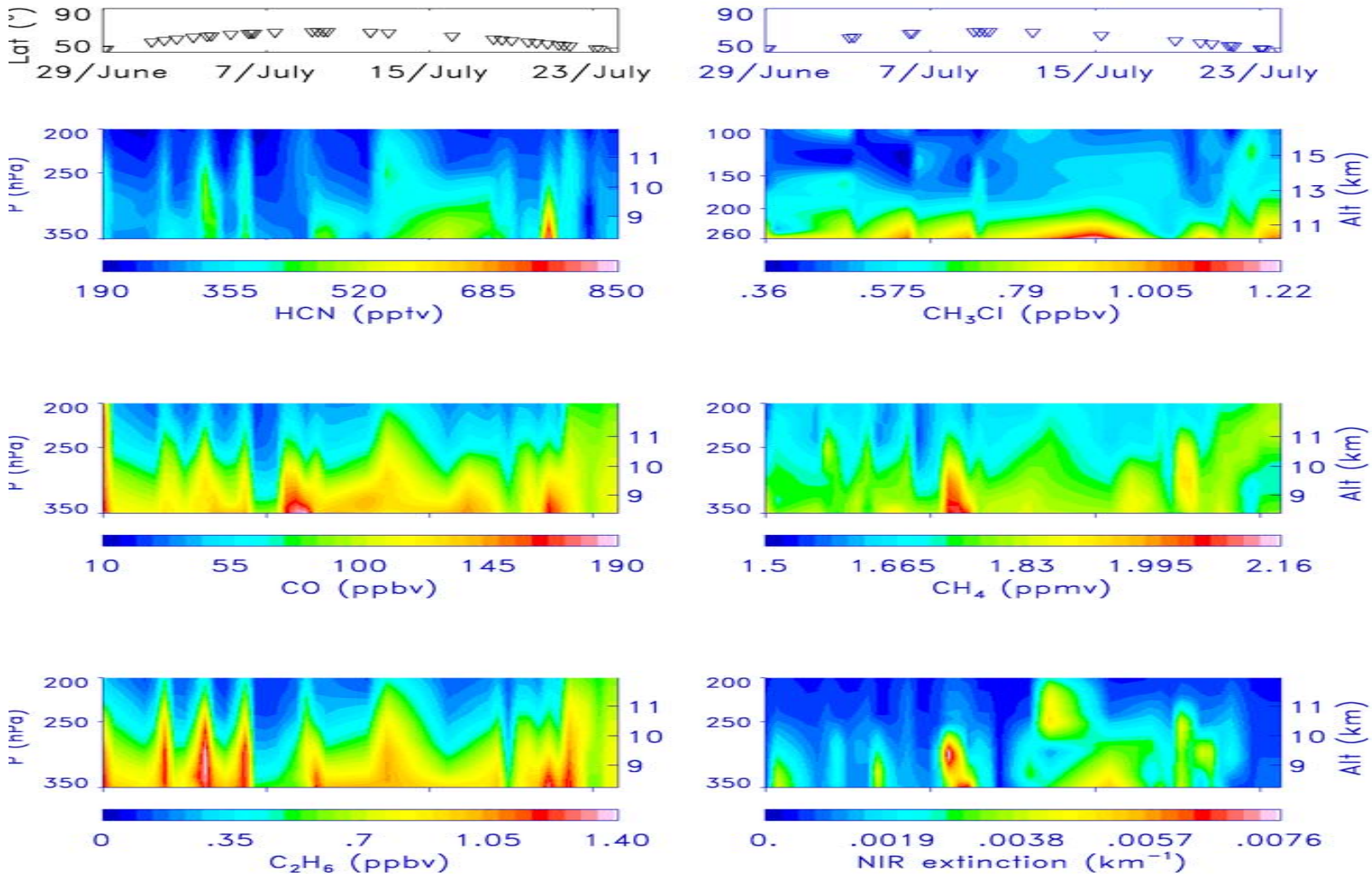
- The Arctic is a particularly sensitive region to global climate change. Both observations and models indicate that as the climate warms, the Arctic warms the most and the fastest
- Boreal wildland fires are an important biomass burning emission source of trace gases including CO_2 and aerosols with large spatial and temporal variability
- Boreal and temporal forests in North America account for more than 15% of the world's forests
- Changes in tundra, boreal forests, and permafrost could lead to changes in emission rates that coincide with the observed and predicted climate warming at mid- to high latitudes
- Large and intense burning liberate huge amounts of carbon, but error in the amount of carbon released from forest fires is one of the major uncertainties in understanding and closing the global carbon cycle budget

  2004 was the worst fire season in Alaska and western Canada on record
- More than 5×10^6 hectares burned
- Total CO emissions during June to August were of comparable order of magnitude to those of the entire continental U.S. for the same time period with CO plumes as high as 600 ppbv (10^{-9} per unit volume) measured *in situ* at 400 hPa during an aircraft flight near Newfoundland

ACE Boreal 2004 Measurements

- ACE measurements recorded between June 29 and July 23, 2004 show mixing ratios up to 189 ppbv for CO, 992 pptv for HCN, 1.09 ppbv for C_2H_6, 3.63 ppbv for CH_3Cl, and 2.09 ppmv for CH_4 in the upper troposphere
- Correlated temporal and spatial variations reflect their common origin and transport and back trajectory calculations suggest the elevated levels originated from biomass burning regions in Alaska or Siberia with transport to the upper troposphere
- ACE CH_3Cl measurements are limited to a minimum altitude of 9 km because of interferences, but they provide the only space-based global tropospheric profile measurements of that molecule
- Correlation of ACE mixing ratios with aerosol extinction provides evidence for particle emissions from the fires
- HCN is a key indicator of fire activity though not measured by TES and MLS measurements are limited to weekly stratospheric zonal means above 30 hPa (~24 km)

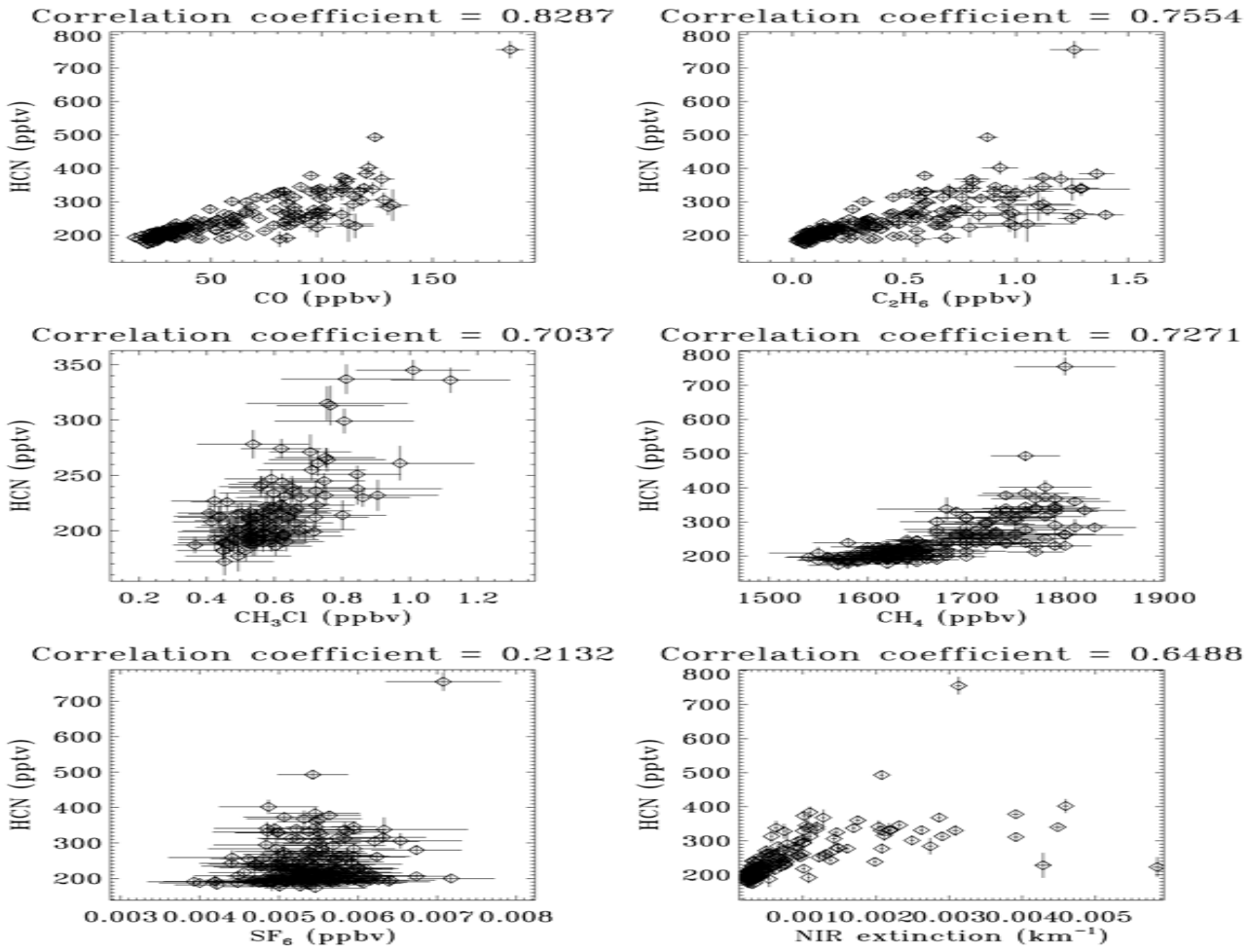
ACE Arctic 2004 Time Series



Measurement of Pyro-convective Plumes

- Elevated upper tropospheric mixing ratios for CO, C_2H_6, HCN, C_2H_6, HCN, and 1.02 μm extinction were measured
- ACE measurements provide an indication of high altitude injection of the fire emissions associated with pyro-convective events in mid-July 2004, an hypotheses supported by aircraft and model analysis

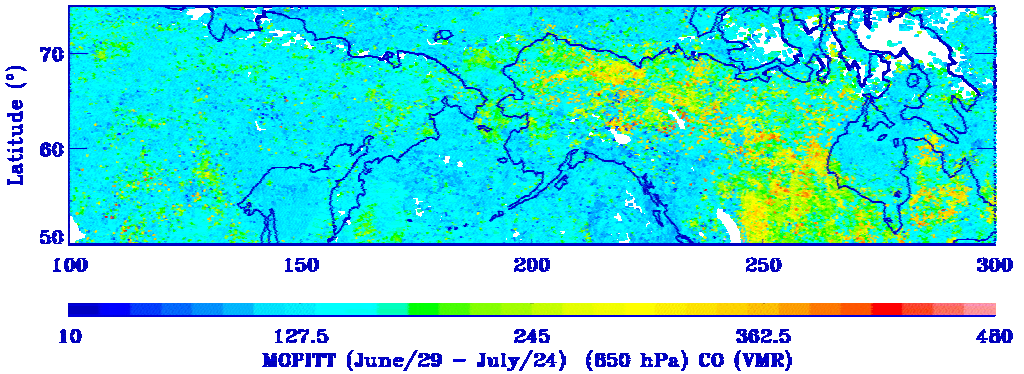
Boreal Upper Tropospheric Correlations with HCN

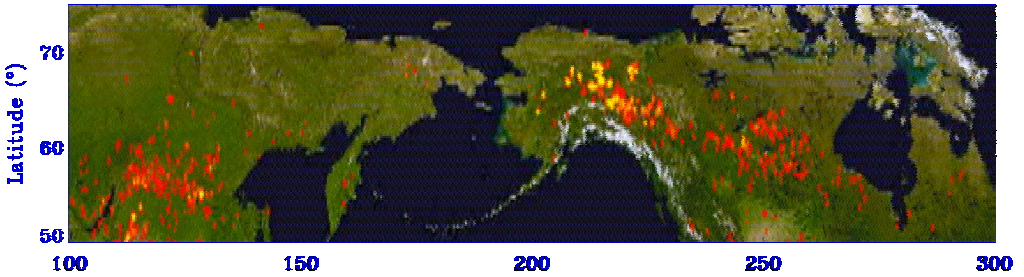


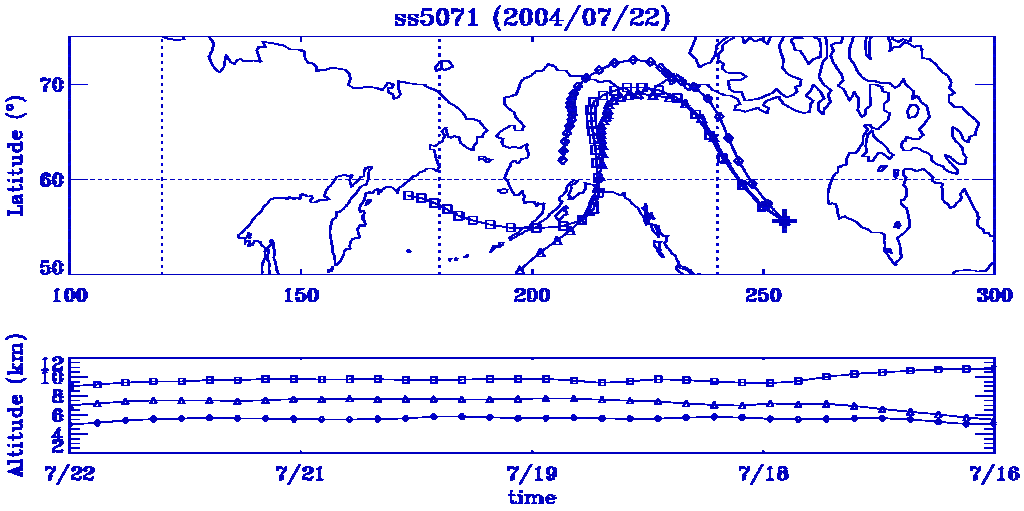
Back trajectories

- Kinematic back trajectories were run with the HYSPLIT 4 model for the locations of several ACE occultations with elevated mixing ratios for biomass burning products at the and altitudes above and below the measured peak mixing ratios
- Results indicate the emissions resulted from lower altitudes, though the scatter in the results makes it difficult to pinpoint the origin of the emissions

MOPITT/MODIS Verification and Back Trajectories







Interpretation

- Assuming the emissions originated from convective events, the varying correlation coefficients from the mixing ratio measurements likely reflect differences in the lifetimes in the Arctic with the lowest correlation measured for HCN vs. CH_3Cl with a lifetime that is longer than for the other molecules except CH_4
- Correlation of the mixing ratios with those from the 1.02 μm extinction likely reflect impact of aerosol emissions
- Measurement of elevated CO is consistent with those from MOPITT, the elevated fire counts from MODIS, and back trajectory calculations which suggest the emissions likely originated from burning regions in Alaska or Siberia with convective transport from the surface to the upper troposphere

ACE Boreal 2004 Result Summary

- ACE measurements recorded between June 29 and July 23, 2004 show mixing ratios up to 189 ppbv for CO, 992 pptv for HCN, 1.09 ppbv for C_2H_6, 3.63 ppbv for CH_3Cl, and 2.09 ppmv for CH_4 in the upper troposphere
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Summary and Conclusions

- ACE continues to provide a wealth of high precision measurements of the upper troposphere to lower thermosphere providing measurements of with more than 30 data products including temperature providing high precision measurements for a wide range of studies such as air quality measurements, trend quantification and verification of international protocols
- Profile measurements provide tests of the precision and accuracy of current spectroscopic parameters
- Key measurements are provided for validation of spaceborne instruments (e.g. MIPAS, SCIAMACHY, Aura) and simultaneous observations for climate change studies
  - Potential for ACE measurements for POLARCAT, an international ground/ship/aircraft atmchem mission over the Arctic in winter/spring/summer 2008 under the auspices of the International Polar Year